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(54) Title: BLENDS OF POLYAMIDES WITH GRAFT COMPOUNDS		
(57) Abstract Polyamide containing polymeric blends and process for their preparation are disclosed. These blends possess improved impact strength and resistance to moisture absorption. Preferred formulations of these polymeric blends comprise: (a) from about 10 to about 90weight percent of a polyamide and from about 90 to about 10 weight percent of a maleic anhydride or acrylic acid grafted isotacticmaleic anhydride or acrylic acid grafted isotactic polypropylene containing polymer; and (b) from about 50 to about 80 weight percent of a polyamide, from about 10 to about 40 weight percent of a maleic anhydride or acrylic acid grafted isotactic polypropylene containing polymer and from about 10 to 25 weight percent of an impact modifying agent, preferably of a maleic anhydride or acrylic grafted ethylene-propylene containing elastomer. For formulation (b), a mixture of a maleic anhydride grafted isotactic polypropylene containing polymer and maleic anhydride grafted ethylene-propylene containing elastomer in a weight ratio of from about 2:3 to about 5:2, respectively and most preferably 3:2, is particularly advantageous. The polyamide containing polymeric blends of the present invention are useful in the fabrication of articles requiring relatively high impact strengths and improved resistance to moisture.		

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BLEND OF POLYAMIDES WITH GRAFT COMPOUNDSBackground of the Invention5 1. Field of the Invention

The present invention relates generally to polyamide containing polymeric blends, and more particularly, but not by way of limitation, to a melt compounded blend of a polyamide and either a graft compound of a graft isotactic
10 polypropylene containing polymer or a graft compound mixture of the graft isotactic polypropylene containing polymer and a graft ethylene-propylene containing elastomer, as well as to a process for producing same. Particularly preferred graft compounds include maleic anhydride grafted or acrylic acid
15 grafted isotactic polypropylene and maleic anhydride grafted or acrylic acid grafted ethylene propylene elastomer, and mixtures thereof.

20 2. Brief Description of the Prior Art

Polymeric resins have long been known for their chemical and physical properties. Molded or extruded resins have found numerous applications, such as in appliances, consumer products, electronics, machine components, automotive parts and the like. However, the physical and chemical
25 properties of the polymeric resins, and thus the components or articles fabricated therefrom, vary widely depending upon the chemical structure of the main chain or backbone of the polymeric resins, as well as the molecular weight of such polymeric resins.

For example, polyamides (i.e. the nylons) are known
30 to be chemically resistant to a large number of solvents, and to have a desired degree of toughness and abrasion resistance. However, the polyamides also possess certain inherent disadvantages such as an affinity to absorb moisture, and a

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relatively low impact strength, unless modified.

In order to modify the properties of polyamides, various polymeric resins have been blended with a polyamide. However, in certain instances mixtures or blends of the poly-
5 meric resins and polyamides have proven unacceptable because the polymeric resins are not compatible with the polyamides, or require special compounding procedures.

The prior art is replete with various methods and toughening agents for improving the impact strength of poly-
10 amides, and thus articles molded or extruded therefrom. For example, it has heretofore been proposed that the impact strength of polyamides can be improved by the incorporation of low molecular weight plasticizers into the polyamides. It has also been proposed that copolymers of ethylene and alpha,
15 beta unsaturated acids or their derivatives be blended with or incorporated into polyamides to improve the impact strength of polyamides; and that graft copolymers prepared from ethylene containing copolymers be incorporated into the polyamides to provide compositions having improved impact
20 strength.

While much effort has heretofore been expended to develop and formulate polymeric blends containing polyamides having an improved impact strength, the inherent problem of moisture absorption has been neglected and often believed to
25 be important as a conditioning component of the polyamides in order for the polyamides to obtain desired impact strengths. Therefore, it would be highly desirable and a significant advance in the art if a polymeric blend containing a poly-
amide could be formulated in which the components were
30 compatible, which readily improved the impact strength of the polyamide and which substantially reduced the affinity of the polyamide to absorb moisture. It is to such a polymeric blend and a process for producing same that the subject invention is directed.

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Summary of the Invention

An object of the present invention is to provide substantially homogeneous polyamide containing polymeric compositions having improved impact strengths and resistance to moisture absorption.

Another object of the present invention, while achieving the before stated object, is to provide an economical process, using conventional equipment, for producing polymeric blends containing polyamides.

Other objects, advantages and features of the present invention will become clear from the following detailed description when read in conjunction with the appended claims.

Accordingly, the present invention provides polyamide containing polymeric blends having improved impact strengths and resistance to moisture absorption. In another aspect, the present invention relates to a process for producing such polyamide containing polymeric blends.

The polymeric blends of the present invention which possess improved impact strengths and resistance to moisture absorption contain: (a) from about 10 to about 90 and preferably 35 to 65 weight percent of a polyamide, preferably having a molecular weight of at least about 2,000, such as nylons; and (b) from about 90 to about 10 and preferably 65 to 35 weight percent of a graft compound or mixture. In one embodiment, the graft compound comprises an isotactic polypropylene containing polymer which is modified by grafting an alpha-beta unsaturated carboxylic acid or its corresponding anhydride onto the polypropylene containing polymer. In another embodiment, the graft compound includes a mixture of the graft polypropylene containing polymer with an ethylene-propylene containing elastomer which is modified by grafting an alpha-beta unsaturated carboxylic acid or its

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corresponding anhydride onto the ethylene-propylene containing elastomer. Thus, the graft mixture generally includes from about 20 to 100 weight percent of the graft polypropylene containing polymer and from about 80 to 0 weight percent of the graft ethylene-propylene containing elastomer. For either graft compound, the preferred components for grafting include from about 0.1 to 3% by weight of maleic anhydride or from about 0.1 to 10% by weight of acrylic acid.

When the mixture of two graft compounds are used with the polyamide, the preferred relative proportions of each component are approximately as follows: from about 50 to 80 percent by weight of the polyamide resin, from about 10 to about 40 percent by weight of the graft polypropylene containing polymer, and from about 10 to about 25 percent by weight of the graft ethylene-propylene containing elastomer, with each graft compound preferably present in the graft mixture in a weight ratio of from about 2:3 to about 5:2, respectively.

The present invention also provides a process for improving the impact strength and resistance to moisture absorption of polyamides. Broadly, the process comprises: (a) admixing from about 10 to about 90 weight percent of particulate polyamide, preferably having a molecular weight of at least about 2,000 with from about 90 to about 10 weight percent of one of the graft compounds described above in particulate form for an effective period of time and at low shear conditions to form a substantially uniform polyamide containing mixture; and, (b) introducing the polyamide containing mixture into a compounding extruder and extruding a polyamide containing heat plasticized, substantially homogeneous mass to form a polyamide containing extrudate of the polyamide and graft compound or mixture. The compounding extruder is operated at a temperature of from about 350°F to about 550°F, at a pressure of from about 500 psi to about

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1,500 psi, and under shear conditions effective to provide substantially uniform mixing of the constituents within the compounding extruder.

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Description

The term "blend" as used herein is to be understood to signify a substantially homogeneous mass of materials, and the resulting extrudate, preferably in pellet form, which is obtained by heating such materials to the melting or softening point under high shear conditions in an extruder.

The polyamide containing polymeric compositions of the present invention, and which unexpectedly possess improved impact strengths and resistance to moisture, are a blend of certain constituents, most preferably, a polyamide, a maleic anhydride grafted isotactic polypropylene containing polymer, and, optionally, a maleic anhydride grafted ethylene-propylene containing elastomer. Thus, the polyamide containing polymeric compositions, which are blends of the these constituents, in one embodiment include from about 10 to about 90 and preferably 35 to 65 weight percent of a polyamide and from about 90 to about 10 and preferably 65 to 35 weight percent of a graft compound or mixture. The most preferred graft compound is an isotactic polypropylene containing polymer to which has been grafted an alpha-beta unsaturated carboxylic acid or anhydride derivative thereof.

25

The term "isotactic polypropylene containing polymer" is used to designate crystalline polypropylene or random and/or block copolymers of polypropylene with ethylene. As one skilled in the art would realize, such random or block copolymers generally include less than about seven percent by weight ethylene. Also, the crystalline form of these polymers clearly designates those having a defined melting point with generally high heat resistance on the order of about 330°F. Also, the isotactic form of these

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polymers, being crystalline in nature, is clearly distinguishable from the syndiotactic or atactic forms, each of which is amorphous and waxy in nature.

The polymeric blends of polyamides with graft isotactic polypropylene containing polymers provide unexpected properties with regard to improved resistance to moisture absorption of the resulting blend. The lower end of the claimed range for this component, i.e., 10% or above, imparts this property to the blend. When the higher end of the range is used, i.e., up to 90%, blends having improved heat distortion properties are obtained.

Another preferred formulation comprises from about from 50 to about 80 weight percent of a polyamide, and from about 10 to about 40 weight percent of the maleic anhydride grafted isotactic polypropylene containing polymer and from about 10 to 25 weight percent of a maleic anhydride grafted ethylene-propylene elastomer.

The relative amounts of the maleic anhydride grafted isotactic polypropylene containing polymer and the maleic anhydride grafted ethylene-propylene elastomer present in the polymeric blend can vary widely. However, it is advantageous for the maleic anhydride grafted isotactic polypropylene containing polymer and the maleic anhydride grafted ethylene-propylene elastomer to be present in a weight ratio of from about 2:3 to about 5:2, respectively, and most desirably, in a weight ratio of about 3:2.

The term "ethylene-propylene containing elastomer" is used to designate ethylene-propylene containing rubber-like compositions which are noncrystalline in structure and characterized as having high flexibility. Preferred ethylene-propylene containing elastomers include ethylene-propylene rubber and ethylene-propylene diene monomer rubber (i.e., EPDM). These compounds are clearly distinguishable from conventional ethylene-propylene polymers or co-polymers

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which are crystalline in nature.

Instead of or along with the graft ethylene-propylene containing polymer, other so-called impact modifying agents can be utilized in conjunction with the graft isotactic polypropylene containing polymer and polyamide to form blends according to the invention. Suitable impact modifying agents include those disclosed in U.S. Patent No. 4,174,358, the content of which is expressly incorporated herein by reference thereto. These agents are used in the same relative amounts as the graft ethylene propylene containing elastomer.

Although maleic anhydride grafted compounds are the most preferred for use in forming the graft compounds, acrylic acid has also been found to be successful. Thus, it appears that any alpha-beta unsaturated acid or anhydride derivatives thereof can be used for preparing such graft compounds. When acids, such as acrylic acid, are used to form the graft compound, the relative amount of acid preferably ranges from 0.1 to 10 weight percent, while for anhydride derivatives, such as maleic anhydride, the amount to be used ranges from about 0.1 to 3 weight percent.

The polyamides which can be employed in the formulation of the polyamide containing polymeric compositions having improved impact strengths and resistance to moisture absorption are well known in the art and embrace those semi-crystalline and amorphous resins, preferably having a molecular weight of at least about 2,000, commonly referred to as nylons. Desirably, the polyamides will be linear polyamides having a molecular weight of at least about 5,000, such as polycaprolactam (6 nylon), polyhexamethylene adipamide (66 nylon), polyhexamethylene azelamide (69 nylon), and mixtures thereof.

As previously stated, the blends of the invention contain from about 10 to about 90 weight percent of a

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polyamide and from about 90 to about 10 weight percent of a graft compound of a graft isotactic polypropylene containing polymer or mixture of the graft polypropylene containing polymer with a maleic anhydride grafted ethylene-propylene containing elastomer. However, when a linear polyamide is employed as the polyamide constituent, such as hexamethylene adipamide, polycaprolactam, and mixtures thereof, the polyamide blends of the present invention desirably comprise from about 35 to 65 weight percent of the polyamide and from about 65 to 35 weight percent of the graft compound or graft mixture.

The preferred maleic anhydride grafted isotactic polypropylene containing polymer constituent of the compositions of the present invention is desirably formulated as a maleic anhydride or acrylic acid grafted homopolymer or copolymer of isotactic polypropylene. The homopolymer or copolymer of polypropylene to which maleic anhydride or acrylic acid is grafted is characterized as having a molecular weight of from about 20,000 to about 500,000 and containing greater than 90% H.I., isotactic polypropylene with a melt flow of 2.0 to 20 grams/10 minutes. Examples of homopolymers and copolymers of polypropylene satisfying the before-stated characterization are the polypropylene polymers manufactured by Himont Corporation and marketed under the trade names "Himont 6501", "Himont 6801", and the polypropylene polymers manufactured by El Paso Products Company and marketed under the trade names "Rexene 11S12", "Rexene 17S2", and the like.

As noted above, these crystalline polypropylene polymers are distinguishable from the ethylene-propylene containing elastomers in that the ethylene-propylene elastomers to which maleic anhydride or acrylic acid is grafted are characterized as having a relatively high Mooney

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content.

Examples of suitable ethylene-propylene containing elastomers onto which maleic anhydride or acrylic acid can be grafted for use in the formulation of the polyamide containing blends of the invention are those products produced by Exxon Chemical Company and marketed under the trade names "Vistalon 702", "Vistalon 719", and "Vistalon 606". Other suitable ethylene-propylene elastomers include those which are generally known in the art, as shown by U.S. Patent No. 4,174,358.

The grafting of maleic anhydride to polymeric backbones, such as polypropylene polymers and ethylene-propylene elastomers, to produce grafted products is generally known. However, especially desirable results have been obtained when the grafting of maleic anhydride to the homopolymers and copolymers of polypropylene, and the grafting of maleic anhydride to the ethylene-propylene elastomer is carried out using the following described process.

Preparation of Maleic Anhydride
Grafted Polypropylene and Ethylene-propylene Mixture

1. A homopolymer or copolymer of polypropylene and an ethylene-propylene elastomer are introduced into a low intensity mixer and blended under low shear conditions for an effective period of time to form a substantially uniform mixture of the polypropylene and the ethylene-propylene elastomer. The polypropylene and the ethylene-propylene elastomer, each of which is in a particulate state, such as a powder or granules, are introduced into the low intensity mixer in amounts sufficient to insure that the polypropylene and ethylene-propylene elastomer are present in the mixture in a weight ratio of from about 2:3 to about 5:2, respectively, and more desirably about 3:2.

2. An effective amount of granulated maleic anhydride,

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generally from about 0.2 to about 2 weight percent, and an effective amount of a catalyst, generally from about 0.02 to about 0.3 weight percent, are introduced into the low intensity mixer containing the substantially uniform mixture of the polypropylene and the ethylene-propylene elastomer and blended under low shear condition for an effective period of time to insure substantially uniform dispersement of the maleic anhydride and the catalyst throughout the mixture of the polypropylene and the ethylene-propylene elastomer.

Blending time of the maleic anhydride and the catalyst is generally three (3) minutes or less. If desired, and depending upon the scale of operation, a dry masterbath of the maleic anhydride and the catalyst in the polypropylene can be formulated, and thereafter mixed with remaining amounts of the polypropylene and the ethylene-propylene elastomer to insure substantially uniform dispersement of the maleic anhydride and the catalyst throughout the mixture of the polypropylene and the ethylene-propylene elastomer.

Any suitable composition well known in the art can be employed as the catalyst for the polyamide, polypropylene, and ethylene-propylene elastomer system that is cable of generating free radicals effective in grafting maleic anhydride to the polypropylene and the ethylene propylene elastomer. Typical of such catalysts are organic peroxides such as peroxyesters and diacyl peroxides. Representative peroxyesters include t-butyl peroxyacetate, t-butyl peroxyacetate, t-butyl peroxyisobutylate, t-butyl peroxyacetate, t-butyl peroxyneodecanoate, t-butyl peroxybenzoate; t-butyl peroxyoctoate, t-butyl peroxy (2-ethylhexanoate), t-amyl peroxyneodecanoate, cumyl neodecanoate, isobutyl peroxy-pivalate, sec-butyl peroxybenzoate, n-butyl peroxyoctoate, t-butyl peroxy-3, 3,5-trimethylhexanoate, t-butyl peroxy-2-methylbenzoate, 2,5-dimethyl-2,5-bis(2-ethylhexanoylperoxy)hexane, 2,5-dimethyl-2,5-bis(benzoylperoxy) hexane, 2-5-dimethyl-2,5-

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bis(octanylperoxy)hexane, di-t-butyl diperoxyphthalate, t-butyl peroxy maleic acid, t-butyl peroxyiso-propylcarbonate, di(sec-butyl)peroxydicarbonate, bis(4-t-butyl cyclohexyl)peroxydicarbonate, diisopropyl peroxydicarbonate, 5 di(n-propyl)peroxydicarbonate, di(2-ethylhexyl)- peroxydicarbonate, dicyclohexyl peroxydicarbonate, dicetyl peroxydicarbonate and the like. Aliphatic diacyl peroxides including acetyl peroxide, lauroyl peroxide, decanoyl peroxide and isononanoyl peroxide, as well as aromatic diacyl peroxides 10 including benzoyl peroxide, p-chlorobenzyl peroxide and 2,4-dichlorobenzoyl peroxide may be used. However, desirable results have been obtained wherein the catalyst is a hindered phenol marketed by Lucidol Chemical Company under the trademark "LUPERCO 101XL" and having a half life at 119°F of about 15 10 hours.

3. The mixture formulated in step 2 above, and containing polypropylene, ethylene-propylene elastomer, maleic anhydride and the catalyst, is continuously fed to a compounding extruder which functions to initiate the grafting 20 reaction of the maleic anhydride with the polypropylene constituent of the mixture and the ethylene-propylene elastomer constituent of the mixture. The extruder is operated at a temperature of from about 350°F to about 550°F and at a pressure of from about 500 psi to about 1,500 psi (that is, 25 at a sufficient temperature and pressure to melt the polypropylene constituents of the mixture and form a substantially homogeneous molten mass and decompose the organic peroxide catalyst, without incurring decomposition of the polymeric constituents of the mixture.)

30 Single screw or twin screw extruders can be utilized. However, twin screw compounding extruders are preferred because of the higher shear produced on the molten mixture in the extruder. When employing a twin screw compounding extruder, the mixture is starve fed to the

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extruder and the screw speed is independently controlled relative to the output rate. The higher screw speeds provide more intensive mixing of the constituents in the mixture and reduce the particle size of the ethylene-propylene elastomer dispersed throughout the polypropylene phase. Thus, screw speeds of from about 100 rpm to about 300 rpm are desired, and more desirably the screw speed is about 150 rpm.

A temperature profile of lower temperatures, which progressively increase from intake to the die head, is desired to insure that the polymeric constituents (i.e. the polypropylene and maleic anhydride) are melted, that the constituents are uniformly mixed, and that the peroxide catalyst is decomposed. Thus, a typical temperature profile of the compounding extruder is as follows:

15	<u>Feed</u>	<u>Transition</u>	<u>Metering</u>	<u>Die Head</u>
	350°F	380°F	450°F	480°F

Desirable results have been obtained wherein the melt temperature of the polymeric constituents in the extruder is maintained at about 450°F, and the extruder is operated at a production rate of about 200 lbs/hr. with a resulting residence time in the extruder of about three (3) minutes. Further, the extruder can be provided with a vent section to devolatilize excess monomer. However, if the constituents of the mixture are controlled within the ratios hereinbefore set forth, it will not generally be necessary to devolatilize excess monomer.

4. The molten grafted composition (which consists essentially of a blend of maleic anhydride grafted polypropylene and maleic anhydride grafted ethylene-propylene elastomer) is extruded through the die head into strands, and thereafter chopped into pellets. The pelletizing of the extrudate can be accomplished by any means well known in the art. For example, the extrudate can be first cooled in a

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waterbath and thereafter passed through a pelletizer; or the extrudate can be cut into pellets at the die head face, such as with a Gala pelletizer.

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Preparation of Polyamide Containing
Polymeric Compositions

1. The pelletized product of the maleic anhydride grafted polypropylene and the maleic anhydride grafted ethylene-propylene elastomer blend, particulate polyamide
10 having a molecular weight of at least about 2,000, and an effective minor amount of an antioxidant capable of stabilizing the maleic anhydride grafted constituent, typically from about 0.05 to about 0.2 weight percent, are introduced into a low intensity mixer, such as a ribbon blender, and blended
15 under low shear conditions for an effective period of time to form a substantially uniform mixture of the maleic anhydride grafted polymeric blend and the polyamide. The pelletized product (i.e. the maleic anhydride grafted polypropylene and the maleic anhydride grafted ethylene-propylene elastomer
20 blend) and the particulate polyamide are introduced into the low intensity mixer in an amount sufficient to insure that the pelletized product and the polyamide are present in the mixture in a weight ratio of from about 1:10 to about 10:1, respectively, and more desirably about 1:1.8. Blending time
25 of the pelletized product, the polyamide and the antioxidant in the low intensity mixer, such as a ribbon blender, is generally for about three (3) minutes or less.

Any suitable composition antioxidant well known in the art can be employed to stabilize the maleic anhydride grafted polymeric blend. Typical of such antioxidant are
30 "Irganox 1010" and "Irganox 1076" marketed by Ciba Geigy Company, and "BHT" (butylated hydroxy toluene) marketed by MNBV Shell Chemical Company, and the like.

2. The resulting mixture of the maleic anhydride grafted
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ted polymeric blend, the polyamide and the antioxidant is continuously fed to a compounding extruder for melt compounding. The extruder is operated at a temperature of from about 350°F to about 550°F and at a pressure of from about 500 psi to about 1,500 psi (that is, a sufficient temperature and pressure to melt the maleic anhydride grafted polymeric blend and the polyamide and form a substantially homogeneous molten mass, without incurring decomposition of the polymeric constituents).

Single screw or twin screw extruders can be utilized in the melt compounding of the mixture. However, twin screw compounding extruders are preferred because of the higher shear produced on the molten mixture in the extruder. When employing a twin screw compounding extruder, the mixture is starve fed to the extruder and the screw speed is independently controlled relative to the output rate. The higher screw speeds provide more intensive mixing of the constituents of the mixture. Thus, screw speeds of from about 100 rpm to about 300 rpm are desired, and more desirably the screw speed is about 150 rpm.

A temperature profile of the extruder of temperatures which progressively increase from intake to the die head is desired to insure effective mixing of the multi-component system which melts over a broad temperature range. Thus a melt temperature of about 500°F to 530°F is desired, and a typical temperature profile of the compounding extruder is as follows:

<u>Feed</u>	<u>Transition</u>	<u>Metering</u>	<u>Die head</u>
400°F	475°F	520°F	520°F

Desirable results have been obtained wherein the melt temperature of the multi-component system is maintained at about 520°F, and the extruder is operated at a production rate of about 200 lbs/hr with a residence time in the extruder of about three (3) minutes.

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3. The molten, heat plasticized mass (which consists essentially of a blend of polyamide, and a preblended composition of maleic anhydride grafted polypropylene and maleic anhydride grafted ethylene-propylene elastomer) is
5 extruded through the die head into strands, and thereafter chopped into pellets. The pelletizing of the extrudate can be accomplished by any means well known in the art. For example, the extrudate can be first cooled in a waterbath and thereafter passed through a pelletizer; or the extrudate can
10 be cut into pellets at the die head face, such as with a Gala pelletizer.

The polyamide containing polymeric compositions of the invention may be modified by one or more conventional additives, such as fibrous and particulate fillers and
15 reinforcement components, lubricants and mold release agents, colorants such as dyes and pigments, and the like. When incorporating fibrous or particulate fillers and reinforcement components into the polyamide containing polymeric composition to form reinforced polymeric products, the amount
20 of the filler or reinforcement component admixed with the polymeric composition can vary widely and will depend to a large extent on the end use of the articles molded or extruded from the polyamide containing polymeric composition. Generally, however, the amount of filler or reinforcement
25 component admixed with the polymeric composition (while the polymeric composition is in a molten state in a compounding extruder and at a position upstream of the intake of the extruder to minimize abrasive wear to the extruder and to minimize breakdown of mineral particles) will be from about 5
30 to about 50 weight percent, based on the weight of the polymeric composition.

Any suitable fibrous and particulate fillers and reinforcement components can be used to provide a reinforced polyamide containing polymeric compositions. Typical of such

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components, while are well known in the art, are carbon fibers, glass fibers, amorphous silica, chalk, talcum, calcium carbonate, calcium oxide, silicon oxide and the like.

The polyamide containing polymeric blends of the present invention are useful in the fabrication of numerous articles, such as wheel covers, automotive parts, and gears. That is, the polyamide containing polymeric blends are particularly useful in the fabrication of articles requiring relatively high impact strengths and improved resistance to moisture.

In order to more fully describe the present invention, the following example is set forth. However, it is to be understood that the example is for illustrative purposes only and is not to be construed as limiting the scope of the present invention as defined in the appended claims.

Example I

A series of experiments were conducted to determine certain chemical and physical properties of articles molded of various polymeric blends, including the impact strength and moisture absorption of such articles, and thus the polymeric blends from which such articles were fabricated.

A. Manufacture of Maleic Anhydride Grafted Polymeric Blend.

Polypropylene granules (marketed by Himont Chemical Company under the trademark "Himont" 6801) and EPDM or ethylene-propylene rubber particles (marketed by Exxon under the trademark "EXXON VISTALON" 719) were introduced into a low intensity ribbon blender in an amount sufficient to provide a weight ratio of the polypropylene to the ethylene-propylene rubber of about 3:2. To this mixture was added 1 weight percent of granulated maleic anhydride and .045 weight percent of a hindered phenol catalyst having a half life at 119°C of about 10 hours (i.e. "Luperco 101XL" marketed by Lucidol Chemical Company). The above constituents were dry blended in the ribbon blender for 3 minutes to insure that a substantially uniform mixture was obtained.

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The dry blended mixture was then fed to a twin screw compounding extruder (i.e. a Werner Pfleider WP ZSK 83 extruder) to initiate a grafting reaction of maleic anhydride on the polypropylene chain and the ethylene-propylene rubber chain. The compounding extruder was operated at approximately 1000 psi, had a production rate of 200 lbs/hr. and a residence time in the extruder of approximately 3 minutes. The screw speed, which was independently controlled relative to the output rate of the extruder, was 150 RPM to insure that a high shear was produced on the molten polymeric mass in the extruder and to provide more intensive dispersion of the rubber particles in the molten polypropylene phase, as well as reducing the particle size of the ethylene-propylene rubber. The molten polymer mass or blend, which was a substantially homogeneous mass, was extruded through the die head of the compounding extruder in the form of strands. The strands were passed through a waterbath to cool the strands, and thereafter the strands were cut into pellets with a Cumberland pelletizer.

	<u>Feed</u>	<u>Transition</u>	<u>Metering</u>	<u>Die head</u>
20	350°F	380°F	450°F	480°F

The pellets obtained were then blended with a polyamide in accordance with the following procedure.

B. Manufacture of Polyamide Containing Blend.

The pellets produced in Section A above (i.e. pellets of a maleic anhydride grafted polypropylene and maleic anhydride grafted ethylene-propylene rubber blend), and particulate polyamide (nylon 6/6 marketed by Dupont Chemical Company under the Trademark "ZYTEL 101") were introduced into a low intensity ribbon blender in an amount sufficient to provide a weight ratio of the pelletize maleic anhydride grafted blend of polypropylene and ethylene-propylene rubber to the polyamide of about 1:1.8. To this mixture was added about 0.1 weight percent of an antioxidant so as to stabilize the maleic anhydride grafted blend. The anti-

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oxidant employed was "Irganox 1010" marketed by Ciba Geigy Company. The above constituents were dry blended in the ribbon blender for 3 minutes to insure that a substantially uniform mixture was obtained.

5 The dry blended mixture was then starve fed to a twin screw compounding extruder (i.e. a Werner Pfleider WP ZSK 83 extruder) which was operated at approximately 1000 psi, had a production rate of 200 lbs/hr. and a residence time in the extruder of approximately 3 minutes. The screw
10 speed, which was independently controlled relative to the output rate of the extruder, was 150 RPM to insure that a high shear was produced on the molten polymeric mass in the extruder and to provide more intensive mixing of the constituents. The molten polymer mass or blend, which was a
15 substantially homogeneous mass, was extruded through the die head of the compounding extruder in the form of strands. The strands were passed through a waterbath to cool the strands, and thereafter the strands were cut into pellets with a Cumberland pelletizer.

20 A typical temperature profile of the compounding extruder, which progressively increases from intake to the die head so as to insure proper melting and mixing of the polymeric constituents and form a substantially homogeneous blend thereof, is as follows:

25	<u>Feed</u>	<u>Transition</u>	<u>Metering</u>	<u>Die Head</u>
	400°F	475°F	520°F	520°F

The pellets so produced, and consisting essentially of a blend of the polyamide and the grafted blend of maleic anhydride grafted polypropylene and maleic anhydride grafted
30 ethylene-propylene rubber, were then used to mold articles. Tests were then conducted to determine the chemical and physical properties of the articles fabricated from the above-defined pellets and polymers of the different compositions. The following Table I is a compilation of the test
35 data from this Example.

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TABLE I

Constituents	Control			No. 4	Product			
	No. 1	No. 2	No. 3		No. 5	No. 6	No. 7	No. 8
Nylon 6	-	70	-	-	60	-	-	-
Nylon 6/6	70	-	-	-	-	55	79	64
Modified Nylon 6/6	-	-	100	-	-	-	-	-
Grafted Polypropylene*	-	-	-	14	-	-	-	-
Grafted Blend**	-	-	-	-	10	5	21	36
Chopped Fiberglass	30	30	-	30	30	-	-	-
Mineral Filler	-	-	-	-	-	40	-	-
Properties								
Flex Modulus (ksi)	970	1,000	245	990	1,000	990	276	270
Tensile Strength (psi)	-	-	7,430	-	-	12,000	7,780	7,560

... cont'd

* Maleic anhydride grafted isotactic polypropylene wherein the polypropylene has an average molecular weight of about 200,000.

** Grafted polymeric blend consisting essentially of three (3) parts by weight maleic anhydride grafted polypropylene and two (2) parts by weight maleic anhydride grafted ethylene-propylene rubber prepared by Procedure A herein.

TABLE I (continued)

Properties	Control				Product			
	No. 1	No. 2	No. 3	No. 4	No. 5	No. 6	No. 7	No. 8
Elongation (%)	-	-	-	-	-	2.0	3.0	3.0
Heat Deflection Temp. (°F)	470	410	-	474	400	220	-	-
Izod Impact-Notched (Ft. lbs/in)	2.2	2.0	18.6	1.7	2.5	1.9	8.5	19.6
Gardner Impact (in-lbs)	-	-	-	100	150	>320	-	-
Moisture Absorption (%) 24 hr. Immersion	1.2	1.5	1.2	0.88	0.9	0.6	1.0	0.7
Moisture Absorption at Equilibrium (%)	-	-	7.2	-	-	-	6.7	4.0

Note: (1) All testing done on dry as molded test bars

(2) Modified Nylon 6/6 is Dupont ST 801, an impact modified nylon 6/6 with maleated EPDM rubber.

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TABLE II

<u>Constituents</u>	<u>Control No. 1</u>	<u>Control No. 2</u>	<u>Formula No. 3</u>	<u>Formula No. 4</u>	<u>Formula No. 5</u>
PolyBond 64-111	70	-	-	22.5	45
Himont PC-072	-	70	45	22.5	-
Nylon 6	-	-	25	25	25
Fiberglass	30	30	30	30	30
Irganox 1076	0.1	0.1	0.1	0.1	0.1
DSTDP	0.2	0.2	0.2	0.2	0.2
<u>Properties</u>					
Tensile Strength (Psi)	11,200	12,400	10,900	12,900	12,800
Flex Strength (Psi)	17,000	15,200	15,900	18,400	19,800
Elongation (%)	4	4	3	4	5
1/8" Notched Izod (ft. lb/in)	2.3	2.2	1.6	1.7	1.8
1/4" Notched Izod (ft. lb/in)	1.8	1.5	1.1	1.3	1.3

... cont'd

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TABLE II (continued)

<u>Properties</u>	<u>Control No. 1</u>	<u>Control No. 2</u>	<u>Form No. 3</u>	<u>Form No. 4</u>	<u>Form No. 5</u>
Gardner Impact (in-lbs)	95	64	48	60	80
HDT @ 264 psi (°F)	290	290	304	300	290

PolyBond 64-111 is an acrylic acid grafted isotactic polypropylene

Himont AC-072 is a maleic anhydride grafted isotactic polypropylene

Irganox 1076 is a hindered phenol stabilizer

DSTDP is disteryl thiodipropionate, another stabilizer

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The above data in Table I clearly indicates the improved impact strength and resistance to moisture absorption imparted to polyamide containing blends wherein such blends consist essentially of a polyamide, a maleic anhydride grafted polypropylene and maleic anhydride grafted ethylene-propylene elastomer. Further, the unique combination of the constituents forming the blends of the present invention results in unexpected and desirable properties being imparted to the polyamide containing blends.

Also, the above data in Table II, which was carried out using acrylic acid instead of maleic anhydride and solely with a graft isotactic polypropylene polymer, further illustrates how to achieve improved resistance to moisture and high heat distortion temperatures in the disclosed blends. These blends were prepared in essentially the same manner as disclosed above except that the effective amount of acrylic acid ranges from about 0.2 to 5 weight percent.

It should also be noted that the manufacturers of certain polypropylene resins already include an antioxidant or stabilizer compound in their formulations. In this invention, however, additional stabilizer is added to the blends to assure that between about 0.05 and 0.2 percent by weight is present so that optimum results are obtained.

It is clear that the present invention is well adapted to carry out the objects and to attain the ends and advantages mentioned herein, as well as those inherent within the present invention. While preferred embodiments of the invention has been described for the purpose of this disclosure, numerous changes can be made which will readily suggest themselves to those skilled in the art and which are encompassed within the spirit and scope of the invention disclosed herein and as defined in the appended claims.

CLAIMS

What is claimed is:

1. A polymeric blend having improved resistance to moisture absorption consisting essentially of:

5 from about 10 to about 90 weight percent of a poly-
amide; and

from about 90 to about 10 weight percent of a graft isotactic polypropylene containing polymer.

10 2. The blend of claim 1 wherein the graft isotactic polypropylene containing polymer is prepared by grafting an alpha-beta unsaturated carboxylic acid or anhydride derivative thereof onto an isotactic polypropylene containing polymer.

15 3. The blend of claim 2 wherein said graft isotactic polypropylene containing polymer is prepared by grafting maleic anhydride or acrylic acid onto isotactic polypropylene or a random or block copolymer of polypropylene and ethylene.

20 4. The blend of claim 1 further comprising an impact modifying agent in an amount sufficient to increase the impact strength of the blend.

25 5. The blend of claim 1 further comprising a graft ethylene-propylene containing elastomer in an amount sufficient to increase the impact strength of the blend.

30 6. The blend of claim 5 wherein the graft ethylene-propylene containing elastomer is prepared by grafting an alpha-beta unsaturated carboxylic acid or anhydride derivative thereof onto an ethylene-propylene containing elastomer.

7. The blend of claim 6 wherein the graft ethylene-propylene containing polymer is prepared by grafting maleic anhydride or acrylic acid onto ethylene-propylene rubber or ethylene-propylene diene monomer rubber.

8. The blend of claim 5 wherein the polyamide is present in an amount of from about 50 to about 80 weight percent; and the graft isotactic polypropylene containing polymer is present in an amount of from about 10 to 40 percent; and the graft ethylene-propylene containing elastomer is present in an amount of from about 10 to about 25 weight percent.

9. The polymeric blend of claim 8 wherein said graft isotactic polypropylene containing polymer and said graft ethylene-propylene containing elastomer are present in a weight ratio of from about 2:3 to about 5:2, respectively.

10. The blend of claim 1 wherein said polyamide is present in an amount of from about 35 to about 65 weight percent and said graft isotactic polypropylene containing polymer is present in an amount of from about 65 to about 35 weight percent.

11. The blend of claim 1 wherein said polyamide is hexamethylene adipamide, polycaprolactam or mixtures thereof, each having a molecular weight of at least 2,000.

12. The blend of claim 1 further comprising from about 5 to about 40 weight percent of a filler component.

13. The blend of claim 1 further comprising from about 0.05 to about 0.2 weight percent based on the weight of the polymeric blend of an antioxidant capable of stabilizing the graft components.

14. The blend of claim 1 produced by a process which comprises:

mixing an isotactic polypropylene containing polymer with an alpha-beta unsaturated acid or anhydride derivative thereof and a catalyst to form a first mixture;

5 extruding said first mixture to form a substantially homogeneous mass of a graft isotactic polypropylene containing polymer;

admixing a polyamide with said graft polymer and an antioxidant to form a second mixture; and

10 extruding said second mixture to form said polymeric blend.

15 15. The blend of claim 14 wherein the process further comprises admixing with said isotactic polypropylene containing polymer, an ethylene-propylene containing elastomer to form a substantially uniform admixture of said isotactic polypropylene containing polymer and said ethylene-propylene containing elastomer prior to mixing said admixture with said acid or anhydride.

20 16. The blend of claim 14 wherein said process utilizes the acid in an amount of between about 0.1 and 10 percent by weight of said admixture, or the anhydride in an amount of between 0.1 and 3 percent by weight of said admixture.

25 17. The blend of claim 14 wherein the process further comprises:

mixing an ethylene-propylene containing elastomer with an alpha-beta unsaturated acid and a catalyst to form a third mixture;

30 extruding said third mixture to form a substantially homogeneous mass of a graft ethylene-propylene containing elastomer;

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and admixing said graft elastomer with said graft polypropylene polymer and polyamide to form the second mixture prior to extruding said second mixture.

18. The blend of claim 14 wherein each of said first and second mixtures and said admixture of said process are
5 formed under low shear mixing conditions.

19. The blend of claim 17 wherein said first mixture is extruded at a temperature of from about 350°F to about 550°F and at a pressure of from about 500 psi to about 1,500
10 psi, the blend of claim 17 wherein said second mixture is extruded at a temperature of from about 480°F to about 550°F and at a pressure of from about 500 psi to about 1,500 psi, and said third mixture is extruded at a temperature from about 350°F to about 550°F and at a pressure from about 500
15 psi to about 1500 psi.

20. The blend of claim 14 wherein said process further comprises cutting the formed polymeric blend into pellets.

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